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POSSIBLE NEW ATOMIC RESONANCES *

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In the course of a study, which is to be published elsewhere, of the response of atoms to external fields, we found theoretical evidence for the possibility of the occurrence of new resonances in atoms, which are different in kind from single-particle type excitations. They mark the coherent response of an atom as a whole.

We have studied the response function $R(\boldsymbol{q}, \omega)$ in terms of the wave vector $\boldsymbol{q}$ and frequency ω of the external field. Assuming that the external field acts via the density operator and that the external field is sufficiently weak for the Born approximation to be valid, $R(\boldsymbol{q}, \omega)$ is simply related to the auto-correlation function of the density fluctuations. The poles of $R(\boldsymbol{q}, \omega)$ signify resonance frequencies of the system, while the imaginary part gives the absorption spectrum, which is expressed conveniently in terms of the differential oscillator strenght distribution $g(\omega)$:

$$g(\omega) = \frac{2\omega}{\pi q^2} \operatorname{Im} R(\boldsymbol{q}, \omega). \qquad (1)$$

Eq. (1) obeys the sum rule $\int g(\omega) \mathrm{d}\omega = N$, N being the number of electrons in the system.

We introduce first the Fourier transform with respect to time of the auto-correlation function in a Hartree-Fock picture, which is of the form

$$Q_0(\boldsymbol{x}, \boldsymbol{x}', \omega) = \lim_{\epsilon \to 0} \sum_n \left(\frac{\Phi_n(\boldsymbol{x}) \Phi_n^*(\boldsymbol{x}')}{\omega - \omega_n + \mathrm{i}\epsilon} - \frac{\Phi_n^*(\boldsymbol{x}) \Phi_n(\boldsymbol{x}')}{\omega + \omega_n - \mathrm{i}\epsilon} \right). \qquad (2)$$

$\Phi_n(\boldsymbol{x})$ denotes the product $u_i^*(\boldsymbol{x})\, u_j(\boldsymbol{x})$ of the one-electron wave functions associated with the one-electron states i and j, and n stands for the single particle excitation $i \to j$. We account for the Coulomb interaction between excitations in an approximation corresponding to the time-dependent Hartree picture, in which the corresponding correlation function $Q(\boldsymbol{x}, \boldsymbol{x}', \omega)$ is found by solving the following integral equation, written symbolically as

$$Q = Q_0 + Q_0 \, v \, Q, \qquad (3)$$

where v denotes the static Coulomb interaction. The response function is expressed in terms of $Q(\boldsymbol{x}, \boldsymbol{x}', \omega)$ as follows,

$$R(\boldsymbol{q}, \omega) = \int \mathrm{d}^3x \, \mathrm{d}^3x' \; \mathrm{e}^{-\mathrm{i}\boldsymbol{q} \cdot (\boldsymbol{x} - \boldsymbol{x}')} \, Q(\boldsymbol{x}, \boldsymbol{x}', \omega). \qquad (4)$$

Various approximations to $g(\omega)$ can be found introducing different assumptions of the structure of $Q_0(\boldsymbol{x}, \boldsymbol{x}', \omega)$ and different approximations in solving (3).

Assuming the energy spectrum to be locally that of a uniform electron gas and assuming local momentum conservation in solving (3) leads to the well-known result

$$Q(\boldsymbol{q}, \omega) = \frac{Q_0(\boldsymbol{q}, \omega)}{1 - (4\pi/q^2)\, Q_0(\boldsymbol{q}\omega)}. \qquad (5)$$

From (5) all dielectric properties of the electron gas follow as first derived by Lindhard [1]. The resulting response function

$$R(\boldsymbol{q}, \omega) = \int \mathrm{d}^3x \, \frac{Q_0(\boldsymbol{q}, \omega)}{1 - (4\pi/q^2)\, Q_0(\boldsymbol{q}, \omega)} \qquad (6)$$

shows that in this approximation, atoms respond locally to the wave vector of the external field only. Such approximation, therefore, cannot give information on possible resonances of an atom as a whole.

Approximate solutions retaining features of coherence in the response of different parts of an atom can be constructed, using the Fredholm expansion of $Q(\boldsymbol{x}, \boldsymbol{x}', \omega)$, where the numerator and the denominator are analytic functions in the coupling constant. To lowest order in the interaction, we find a dispersion relation of the form

$$1 - \int \mathrm{d}^3x \, \mathrm{d}^3x' \; Q_0(\boldsymbol{x}, \boldsymbol{x}', \omega) \, (1/|\boldsymbol{x} - \boldsymbol{x}'|) = 0. \qquad (7)$$

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Introducing a schematic Hartree model for the atom in which the matrix elements of the interaction factorises as in the schematic model introduced by Brown [2] for a discussion of giant resonances in nuclei, (7) is indeed the exact dispersion equation.

We indentify the roots of eq. (7), including higher terms in the coupling constant where required, as the resonance frequencies of atomic modes.

The formal derivation of the response function and of $g(\omega)$ for the schematic Hartree model is a straightforward matter and will be discussed elsewhere. Clearly, a detailed analysis of eq. (7) for the spectrum of a real atom is a rather formidable program; work is in progress along these lines.

For a survey of the contents of eq. (7) it is illuminating to recast the theory in terms of the statistical model of the atom, since then the atomic properties only depend parametrically on the atomic number Z. We again take for the local Q_0 the propagator function of the high-density gas. But instead of proceeding to eq. (4) via eq. (5), we sum over the Fredholm expansion to leading terms in Z in each expansion term. The approximate dispersion relation for the statistical atom now becomes.

$$1 + (2\pi)^{-3} \int d^3x\, d^3q \log \{1 - 4\pi Q_0(\boldsymbol{q}, \omega)/q^2\} = 0, \quad (8)$$

taking for the imaginary damping terms only the principal values of the logarithm between $-i\pi$ and $+i\pi$.

To get a first qualitative idea about the nature of these resonances, one may solve eq. (8) approximately by retaining only the important contributions for small $|\boldsymbol{q}|$ while neglecting the contributions for $|\boldsymbol{q}|$ larger than a suitably chosen $|\boldsymbol{q}_c(\boldsymbol{x})|$ where local plasma states decay rapidly into particle-hole states. We set $q_c^3 = \beta(4\pi)^{\frac{1}{2}}\omega_0$, where in atomic units $\omega_0(\boldsymbol{x}) = [4\pi\rho(\boldsymbol{x})]^{\frac{1}{2}}$, and $\rho(\boldsymbol{x})$ the electron density at the point $\boldsymbol{x}$ in the atom, β is a constant ~ 1. We have calculated a solution curve of eq. (8) for a statistical atom based on a simple description of atomic structure due to Bohr [3]. The result is shown in fig. 1. The following notation is used: $\gamma = \omega/\omega_c$, where $\omega_c = K_c Z$ is a maximum frequency in the statistical model, and $\varkappa = \beta/K_c$

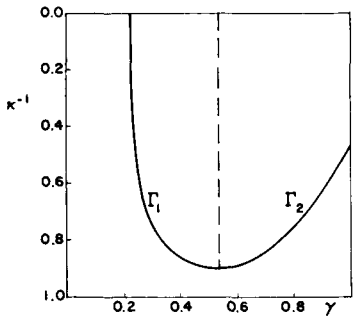

Fig. 1. Solution curve for eq. (8) in the Bohr model $\gamma = \omega/\omega_c$, where $\omega_c = K_c Z$ is the maximum frequency. $\varkappa = \beta/K_c$ is a parameter related to the momentum cut-off $q_c = \beta(4\pi)^{\frac{1}{2}}\omega_0$.

is a dimensionless Z-independent parameter of the model. For $\varkappa = 0$, we retrieve eq. (6). The curves show that for $\varkappa$ smaller than a critical value, no root of eq. (8) exists. For $\varkappa$ larger than this critical value (here ~ 1.11), two roots Γ_1 and Γ_2 appear which separate more and more as $\varkappa$ inceases. To the extent that this preliminary calculation for a simple statistical model applies to real atoms, we conclude that the new atomic resonances may appear over a more or less extended intermediate range of atomic frequencies. We will report on the full response function for the statistical Thomas-Fermi atom in a forthcoming publication. If solutions of eq. (7) exist in this case, an estimate can be made of the oscillator strength residing in these resonances, which will indicate the possibility for observing and indentifying the atomic modes where they exist.

References

1) J. Lindhard, Dan. Mat. Fys. Medd. 28 (1954) no. 8
2) G. E. Brown, Lectures on many-body problems (Nordita, Copenhagen, 1961).
3) N. Bohr, Dan. Mat. Fys. Medd. 18 (1948) no. 8, 99. N. Bohr and J. Lindhard, Dan. Mat. Fys. Medd. 28 (1954) no. 7, 9.

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